Highway proximity and black carbon from cookstoves as a risk factor for higher blood pressure in rural China

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Air pollution in China and other parts of Asia poses large health risks and is an important contributor to global climate change. Almost half of Chinese homes use biomass and coal fuels for cooking and heating. China's economic growth and infrastructure development has led to increased emissions from coal-fired power plants and an expanding fleet of motor vehicles. Black carbon (BC) from incomplete biomass and fossil fuel combustion is the most strongly light-absorbing component of particulate matter (PM) air pollution and the second most important climate-forcing human emission. PM composition and sources may also be related to its human health impact. We enrolled 280 women living in a rural area of northwestern Yunnan where biomass fuels are commonly used. We measured their blood pressure, distance from major traffic routes, and daily exposure to BC (pyrolytic biomass combustion), water-soluble organic aerosol (organic aerosol from biomass combustion), and, in a subset, hopane markers (motor vehicle emissions) in winter and summer. BC had the strongest association with systolic blood pressure (SBP) (4.3 mmHg; *P* < 0.001), followed by PM mass and water-soluble organic mass. The effect of BC on SBP was almost three times greater in women living near the highway [6.2 mmHg; 95% confidence interval (CI), 3.6 to 8.9 vs. 2.6 mmHg; 95% CI, 0.1 to 5.2]. Our findings suggest that BC from combustion emissions is more strongly associated with blood pressure than PM mass, and that BC's health effects may be larger among women living near a highway and with greater exposure to motor vehicle emissions.

cardiovascular disease | household air pollution | solid fuels

Particulate matter (PM) air pollution is a leading health risk factor (1) and primary contributor to anthropogenic climate change (2). Air pollution is notoriously high in China and other parts of Asia. China's rising energy demands have led to increased air pollution emissions from coal-fired power plants (3). Its motorized transport growth is the fastest in the world with the number of motor vehicles projected to quadruple in the next two decades, reaching over 380 million by 2030 (4). Meanwhile, nearly half of all Chinese still cook and heat their homes with highly polluting biomass and coal fuels (5). The resulting PM concentrations routinely exceed the World Health Organization's (WHO) annual Air Quality Guideline of 10 $\mu g/m^3$ by a factor of 10 or more (6) and are associated with a number of adverse health outcomes, including cardiovascular diseases (1, 7).

PM differs in chemical properties, size, and possibly effects on human health. Black carbon (BC) and organic carbon PM are emitted during incomplete biomass and fossil fuel combustion and seem to have important effects on both climate and human health. BC affects the regional and global climate by absorbing solar radiation and heating the atmosphere and is the second most important climate-forcing human emission, after carbon dioxide (8). Coemitted organic carbon may further influence radiative forcing by acting as cloud condensation nuclei (9). These specific characteristics and sources of PM may also impact its toxicity to humans (10).

We previously found that daily exposure to PM <2.5 microns in aerodynamic diameter ($PM_{2.5}$) was associated with higher blood pressure in older Chinese women cooking with biomass fuels (11). In the current study, we used chemical and optical methods to analyze the $PM_{2.5}$ exposure samples for BC and organic components and evaluated their associations with blood pressure, the leading risk factor for cardiovascular diseases, worldwide and in China (1). We enrolled 280 women aged 25–90 y who lived in six villages in the surrounding area along the Yunnan–Tibet Highway in the Himalayan foothills of Yunnan Province, China. These women were subsistence farmers and used biomass fuels (largely wood and crop residues) for cooking and space heating. Details about the study site, household fuel and stove use patterns, and exposure to other PM sources like direct and involuntary tobacco smoking are reported elsewhere (12).

The independent and combined effects of different PM components from various sources on human health are poorly understood. We used measurements of women's daily PM exposure and proximity to a highway to examine how PM composition and sources affect the hazardous effects of blood pressure in this group, using proximity to the highway as a proxy for exposure to motor vehicle emissions using organic tracers.

Results

We enrolled 280 women (mean age: 51.9 y), 18% of whom were overweight [body mass index (BMI) = 25–30 kg/m^2] and 4% obese (BMI ≥ 30 kg/m^2). Mean systolic and diastolic blood pressure (SBP and DBP) were 120 [95% confidence interval (CI), 118 to 122] and 72 mmHg (95% CI, 71 to 73), respectively.

Significance

Air pollution is a leading health risk factor and important contributor to regional climate change in China and other parts of Asia. China's particulate matter (PM) air pollution dramatically exceeds health guidelines and is impacted by industrial emissions, motor vehicles, and household use of biomass and coal fuels. Black carbon (BC) from biomass and fossil fuel burning is a major climate-forcing component of PM. We found that BC exposure from biomass smoke is more strongly associated with blood pressure than total PM mass, and that coexposure to motor vehicle emissions may strengthen BC's impact. Air pollution mitigation efforts focusing on reducing combustion pollution are likely to have major benefits for climate and human health.

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Thirteen percent (n = 37) of participants were hypertensive, of whom 17% (n = 6) were taking antihypertensive medication. None of the participants reported a previous cardiovascular event, and just two women (<1%) reported physician-diagnosed diabetes (Tables S1 and S2).

Exposure of Rural Chinese Women to $PM_{2.5}$ Mass, BC, Organic Carbon, and Motor Vehicle Emission Tracers. Women's geometric mean 24-h $PM_{2.5}$ exposure in summer was 55 μg/m^3 (range: 9–492) and in winter was 117 μg/m^3 (range: 22–634) (12). These levels greatly exceed the WHO's 24-h Air Quality Guideline of 25 μg/m^3. Geometric mean BC exposure was 5.2 μg/m^3 (4 μg/m^3 in summer and 6 μg/m^3 in winter; range: 2–44) which is lower than BC exposure in central New Delhi traffic (42 μg/m^3) (13) but surpasses daytime ambient BC in Beijing, Mexico City, and several cities in Brazil (range: 1.9–4.8 μg/m^3) (14–16). Exposure to water-soluble organic mass (WSOM), a more specific marker of primary biomass smoke, was considerably higher than BC (geometric mean of 12 μg/m^3 in summer and 33 μg/m^3 in winter; range: 1–352) (Table 1), indicating that women's exposure is greatly influenced by biomass combustion (17). Exposure to all pollutants was lower in the summer compared with the winter season; however, the relative contribution of BC and WSOM fractions to $PM_{2.5}$ mass was approximately the same across seasons and for different age groups (Fig. S1).

The PM components had a low-to-moderate correlation, with higher correlations in summer than in winter (Table S3). The largest correlation was between $PM_{2.5}$ and WSOM (r = 0.67), followed by $PM_{2.5}$ and BC (r = 0.48), and finally BC and WSOM (r = 0.44). The PM–BC correlation for rural Chinese women in our study was lower than in outdoor air pollution studies in North America and Europe and in urban Shanghai (PM–BC correlation range = 0.50–0.90) (18, 19). Similarly, BC and hopanes were barely correlated (r = 0.18) in our study, even though both are emitted from motor vehicles. These correlations were low possibly because women's exposures were dominated by a single emission source, namely biomass combustion, rather than traffic emissions or an urban mixture of sources.

Among women living relatively close to the highway that passed through our study site (i.e., less than the median distance of 208 m in our sample), daily BC exposure was slightly higher than those living farther away in winter, but the opposite occurred in summer (Table S4). Distance from the highway was not strongly related to BC exposure, with each 100 m from the highway associated with a 0.02 ln(μg/m^3) lower BC exposure (95% CI, 0 to 0.02; P = 0.30). In contrast, average exposure to hopanes, specific tracers of motor vehicle exhaust in our study setting, was significantly higher among women living in the village closest to the highway (median distance = 76 m) compared with those in the village farthest from the highway (median distance = 548 m) (4.6 vs. 1.1 ng/m^3; P < 0.001 for both seasons; Table S4). In fact, the average near-highway hopane exposures exceeded occupational levels among US trucking terminal workers (4.6 vs. 1.9 ng/m^3) (20).

Associations with Blood Pressure. We evaluated the associations of SBP and DBP with exposure to $PM_{2.5}$ mass, BC, and WSOM. We express the results as the changes in SBP or DBP associated with a 1-ln(μg/m^3) increase in pollutant exposure using one and two-pollutant multivariate mixed-effects models because there was evidence of a nonlinear association (21, 22).

BC exposure had the largest independent effect on blood pressure among rural Chinese women (Fig. 1). In models with just one pollutant, a 1-ln(μg/m^3) increase in BC was associated with 4.3-mmHg higher SBP (P < 0.001), followed by $PM_{2.5}$ mass (2.2 mmHg; P = 0.002) and WSOM (1.2 mmHg; P = 0.06). The estimated effect of BC on SBP changed little (7% change) and remained statistically significant after including $PM_{2.5}$ mass or WSOM in the model. In contrast, the estimated effect of $PM_{2.5}$ mass on SBP decreased by 77% and was no longer statistically significant when BC exposure was added to the model. WSOM had no effect on SBP after other PM components were included in the model (Fig. 1*A*). We found the same strong and statistically robust relationship between BC exposure and DBP (Fig. 1*B*). Our conclusions remained the same when evaluating the changes in blood pressure associated with an interquartile range (IQR) increment in log-transformed pollution exposures, although the difference in the estimated effect of BC on blood pressure relative to PM and water-soluble organic carbon (WSOC) was slightly reduced. An IQR increase in ln(BC) had the strongest and most robust association with higher SBP (3.6 mmHg; 95% CI, 2.0 to 5.2), followed by ln($PM_{2.5}$) (2.8 mmHg; 95% CI, 1.0 to 4.6) and ln(WSOM) (1.8 mmHg; 95% CI, 0.1 to 3.5) (Fig. S2).

We conducted a separate analysis for younger (25–50 y) vs. older women (>50 y). BC was more strongly associated with blood pressure than PM mass among both younger and older women. A 1-ln(μg/m^3) increase in BC exposure was associated with a 1.8-mmHg (95% CI, 0 to 3.6) higher SBP in younger women at the sample average, compared with no effect for $PM_{2.5}$. Among women >50 y old, a 1-ln(μg/m^3) increase in BC was associated with a 7.4-mmHg (95% CI, 4.0 to 10.8) higher SBP and a 2.9-mmHg (95% CI, 1.1 to 4.7) higher DBP (Fig. 2). Our conclusions did not change when evaluating the changes in blood pressure associated with IQR increases in ln(BC) and ln (PM) exposures by age. The effect of an IQR increase in ln(PM) on blood pressure was similar to models estimating a 1-ln(μg/m^3) change in pollution, while the effect of the IQR change in ln(BC) was slightly reduced. In older women, an IQR increase in ln(BC) was associated with a 6.2-mmHg (95% CI, 3.4 to 9.0) higher SBP and a 2.4-mmHg (95% CI, 0.9 to 3.9) higher DBP. In comparison, an IQR increase in ln(PM) was associated with a 5-mmHg high SBP (95% CI, 1.9 to 8.1) and a 2.2-mmHg (95% CI, 0.5 to 3.9) higher DBP. (Fig. S3).

The relatively consistent pollution ratios across age groups and seasons (Fig. S1) suggest that stronger blood pressure effects of pollution in older women are not a result of age-specific differences in PM composition. Excluding women taking hypertensive medication from the analysis did not change our estimated

Table 1. Descriptive statistics for 24-h average personal exposure to $PM_{2.5}$ mass, BC, and WSOM (μg/m^3) among Chinese women cooking with biomass fuels, by season

	Summer				Winter			
Pollutant	*n* (missing)*	GM (95% CI)	Min–max	IQR	*n* (missing)	GM (95% CI)	Min–max	IQR
$PM_{2.5}$ mass	214 (0)	55 (49 to 62)	9–492	61	262 (0)	117 (107 to 128)	22–634	120
BC	211 (3)	4 (4 to 4)	2–14	2	262 (0)	6 (6 to 7)	2–44	7
WSOM	211 (3)	12 (11 to 14)	1–235	21	261 (1)	33 (30 to 37)	1–352	36

*The sample size (*n*) refers to the 24-h exposure sample for a woman enrolled in our study. Missing values are for filters that were damaged during optical or chemical analysis (~1% of samples). GM, geometric mean; Min–max, minimum to maximum.

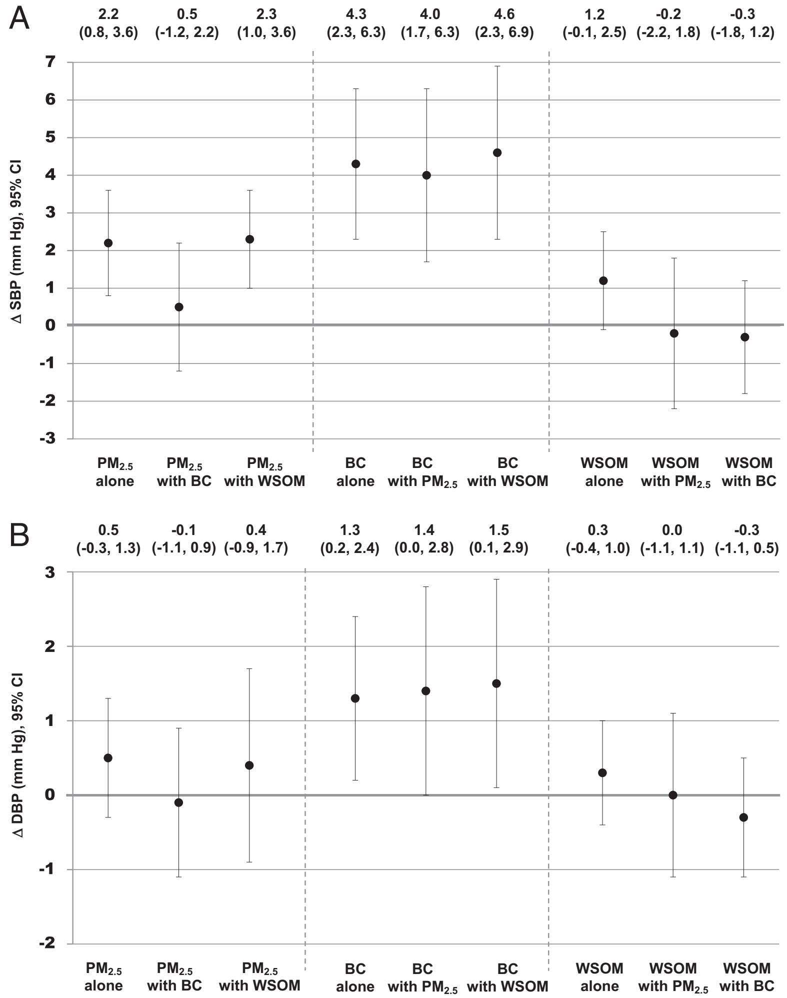

Fig. 1. Associations of personal exposure to PM mass, BC, and WSOM on (*A*) SBP and (*B*) DBP using one- and two-pollutant mixed-effects regression models. ΔSBP or ΔDBP represent the difference in SBP or DBP (with 95% CIs) associated with a 1-ln(μg/m^3) increase in pollutant exposure.

effects of a 1-ln(μg/m^3) increase on SBP (7.3 mmHg; 95% CI, 4.2 to 10.5) or DBP (2.9 mmHg; 95% CI, 1.2 to 4.5) in older women >50 y old.

Distance from Highway, BC Exposure, and Blood Pressure. BC exposure had a larger effect on blood pressure among women living closer (i.e., <208 m from the highway) to the highway that passes through our study site than those living farther away. In the former group, a 1-ln(μg/m^3) increase in BC exposure was associated with almost threefold higher SBP than in women living away from the highway (6.2 mmHg; 95% CI, 3.6 to 8.9 vs. 2.6 mmHg; 95% CI, 0.1 to 5.2; interaction $P = 0.04$). The effect of BC exposure on DBP was also noticeably larger among women living near the highway (2.6 mmHg; 95% CI, 1.0 to 4.2 vs. 0.3 mmHg; 95% CI, −1.3 to 2.0; interaction $P = 0.02$). The 3.6-mmHg larger effect of 1-ln(BC) on SBP among women near the highway is similar to the SBP impact of a modest reduction in sodium intake (−4.2 mmHg per 4.4-g daily reduction) (23) and within the range of an SBP decrease with use of antihypertensive medication (−2.5 to 12 mmHg) (24).

Highway proximity was also a predictor of the effect of BC on blood pressure at the village level, with the effect strongest among women living in villages near the highway and absent among those living in the two villages farthest away from the highway (Fig. 3). Distance from the highway was not independently related to blood pressure, nor did the relationship between $PM_{2.5}$ or WSOM exposure and blood pressure differ by distance from the highway (all interaction *P* values >0.80).

Discussion

We found that BC exposure was more strongly associated with blood pressure than $PM_{2.5}$ mass and WSOM among Chinese women cooking with biomass, and that the effect was stronger among women living near the highway and with greater exposure to motor vehicle emissions. These findings provide several important directions for future health effects studies and for policies and other mitigation strategies aimed at reducing air pollution emissions and exposures.

First, BC may be a useful indicator of the cardiovascular health and climate benefits of interventions that lower air pollution concentrations and exposures. Air pollution mitigation efforts and guidelines, including those in China, have traditionally focused almost exclusively on PM rather than its components or sources (25). However, different interventions may affect PM components, like BC, by varying amounts. For example, some so-called improved cookstoves have emitted higher BC concentrations than traditional stoves, even if they reduced $PM_{2.5}$ mass (26, 27). Not all mitigation options that reduce PM emissions will also reduce total climate forcing or, potentially, benefit health. The inclusion of BC as an outdoor air quality indicator has been proposed (18,

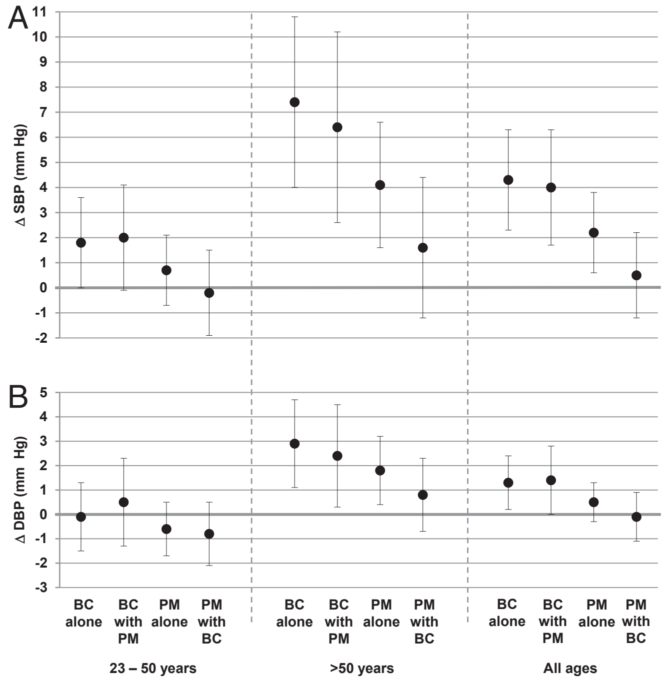

Fig. 2. Associations of personal exposure to $PM_{2.5}$ mass and BC on (*A*) SBP and DBP (*B*) using one- and two-pollutant mixed-effects regression models, by age. ΔSBP or ΔDBP represent the difference in SBP or DBP (with 95% CIs) associated with a 1-ln(μg/m^3) increase in pollutant exposure.

28) but not adopted, and current guidelines for evaluating biomass cookstoves focus on $PM_{2.5}$ and carbon monoxide (29).

Second, BC could be an important exposure assessment tool for future health studies. The larger magnitude of blood pressure response and extension of the health impact to younger women strengthens the importance of our initial finding on $PM_{2.5}$ exposure and blood pressure. Our results support a recent meta-analysis of studies in US and European cities showing that ambient BC concentrations were more strongly associated with cardiovascular mortality and hospital admissions than PM mass (18). If BC is more strongly linked with health than PM, its measurement will facilitate smaller sample sizes and more accurate estimate health impacts of air pollution, and of interventions and policies. There are several methods to measure BC, including simple and low-cost optical assessment on existing PM filter samples or real-time measurement using a new generation of lightweight personal samplers. The inclusion of BC in studies already measuring $PM_{2.5}$ requires less additional effort and resources compared with other combustion markers like WSOM or organic hopanes.

Finally, we found an indication that the cardiovascular effect of BC from biomass smoke may be stronger if there is coexposure to motor vehicle emissions. Our results demonstrate that the blood pressure effect of BC observed in the United States and Europe (30, 31) is not limited to high-income countries where BC is primarily from motor vehicles, although residential biomass combustion contributes to winter ambient air pollution in northern climates (32–34). The stronger health effect of BC from roadway exposure or combined roadway–biomass exposure may also be an important environmental risk factor for cardiovascular diseases in developing countries like China where the number of motor vehicles is rapidly increasing and household use of biomass and coal fuels persists (5, 35).

There are several possible reasons for the stronger effect of BC on blood pressure. One is that BC more closely identifies PM from combustion sources than heterogeneous PM mass does, which comprises particles from all sources, or WSOM, which is both emitted as a primary pollutant and formed as a secondary aerosol. Toxicological studies indicate that PM from incomplete combustion may be more toxic in macrophage and fibroblast cell lines than PM from more complete combustion (36, 37). It is also possible that other components of combustion-related PM contribute to the observed health impacts, with BC acting as a surrogate for their levels. In vitro studies indicate possible toxicity of certain organic constituents in PM from biomass combustion and suggest that BC may be a carrier of these compounds for uptake into macrophages and epithelial cells (36, 38). BC may be operating as an indicator for a larger category of primary combustion particles with varying toxicity to humans, which, in addition to BC, could include metals or polycyclic aromatic hydrocarbons, any of which could act individually or in combination to increase blood pressure (39). Although we cannot determine the single or combination of PM components responsible for the stronger BC effect in our study, our results suggest that a reduction in PM exposure containing BC and other combustion-related particles for which BC is an indicator should lead to a reduction in the adverse health and climate impacts of air pollution.

Our study is limited by its cross-sectional design. However, cooking with biomass is a long-term behavior and all residents have lived in their current homes throughout their adult lifetime. Thus, 24-h PM exposure is a measure that is typical except for seasonal and day-to-day variability; we found little variability in the relative composition of PM exposure by season or daily patterns for women in our study. We considered that factors associated with both blood pressure and highway proximity (e.g., excessive noise or stress from living near the highway) might explain the stronger effect of BC among women closer to the highway. However, highway proximity did not affect the relationship between $PM_{2.5}$ mass and blood pressure, suggesting that distance from the highway is not a proxy for these potentially confounding variables given that $PM_{2.5}$ exposure is also associated with both motor vehicle emissions and blood pressure.

Our results are consistent with several intervention studies in Latin America that found a decrease in SBP (~3–6 mmHg) in older women who switched from a traditional open fire cookstove to a less-polluting chimney stove (40, 41). Although blood pressure is an important risk factor for cardiovascular diseases and overall global burden of disease (1), further research should assess the associations of BC with other health outcomes, including those in vulnerable populations like children. Our study is

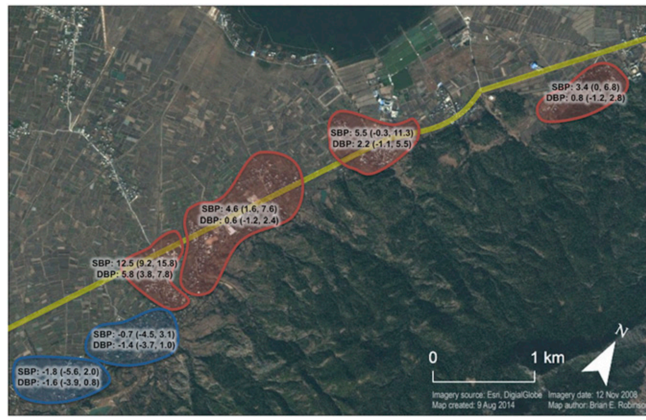

Fig. 3. Associations of personal black carbon exposure and blood pressure (in mm Hg), by village. Estimates and 95% confidence intervals are the changes in SBP and DBP (in mm Hg) associated with a 1-ln(μg/m^3) increase in black carbon exposure, by village. The six villages are outlined in red or blue and the yellow line denotes the Yunnan-Tibet highway. DBP, diastolic blood pressure; SBP, systolic blood pressure.

also limited by proxy measurements, using distance, of motor vehicle exposure in the full sample. Distance from the highway has nonetheless been used as an indicator of roadway air pollution exposure in developed countries (42, 43). Further, women living nearest to the highway had significantly higher exposure to hopanes, direct markers of motor vehicle emissions, than women farthest from the highway in our subsample tracer analysis.

Conclusion

Our results show that the effect of BC exposure on blood pressure is two or more times larger that of $PM_{2.5}$ mass and WSOM among rural Chinese women using biomass fuels. We also found evidence that BC from biomass smoke is associated with higher blood pressure in the presence of motor vehicle emissions as a coexposure. Our findings suggest that BC has direct relevance as an important environmental risk factor for cardiovascular diseases and support the use of BC as a pollution indicator in future health studies and in the evaluation of air pollution mitigation programs. More broadly, our results may be useful in forming policy aimed at reducing air pollution and improving public health in China and other developing countries. China recently committed to spending US$275 billion over the next 5 y to reduce air pollution (44), but targets for new vehicle emission standards are absent from recently announced mitigation plans (45). In addition, China's current air pollution targets and programs focus on PM reductions. The BC reduction achieved with any mitigation strategy is not always proportional to the reduction in PM mass, and our results show that BC may be more strongly associated with health outcomes in addition to warming the climate. As motorized transport and subsequent traffic emissions increase throughout China, air pollution policies and mitigation efforts that focus on BC control might have the largest benefits for climate and human health.

Materials and Methods

We recruited 280 women ≥25 y old between December 2008 and August 2009. None were previous or current tobacco smokers. Families in this region had similar diets, lifestyles, and socioeconomic backgrounds. The Institutional Review Boards at the University of Wisconsin–Madison and Yunnan Provincial Health Bureau approved this research protocol and obtained informed oral consent was obtained from all participants.

Personal Air Pollution Exposure Measurements. We measured the participants' 24-h exposure to $PM_{2.5}$ on 1–3 consecutive sampling days in winter and summer. Participants wore a waistpack holding lightweight air samplers that collected $PM_{2.5}$ on Teflon filters. They were instructed to perform routine daily activities while wearing the waistpack, but could place it within 1 m while sitting or sleeping and within 2 m while bathing. Field staff monitored compliance through home visits.

$PM_{2.5}$ mass was gravimetrically estimated on all exposure samples and blanks using a high-precision microbalance in a temperature and humidity controlled room. They were then analyzed for BC, WSOC, and mobile source tracers at the University of the Chinese Academy of Sciences. We estimated BC concentrations using reflectance analysis with an Optical Transmissometer Data Acquisition System (Model OT21; Magee Scientific) (46).

We used the nonpurgeable organic carbon method described in Timonen et al. (47) to estimate WSOC concentrations. Briefly, filter sections were extracted with high purity water and analyzed with an organic carbon analyzer (Shimadzu Corp.). The resulting WSOC exposures were multiplied by 2.0 to yield WSOM from biomass burning (48, 49). For exposure samples from women in the village either nearest to ($n = 32$ women) or farthest from the highway ($n = 53$ women), we analyzed the remaining filter sections for hopanes, nonpolar organic tracers of motor vehicle exhaust, using the extraction-derivatization method with GC/MS described by Zhang et al. (50).

Details about these PM components, their measurement, and related quality assurance and control practices are described in Baumgartner et al. (12) and discussed in *SI Text*.

Physiological and Health Parameters. Initial questionnaires evaluated household demographics, socioeconomic status, secondhand-smoking status, and medical history. We conducted in-home blood pressure measurements using an automated device and following standard recommendations (51). We recorded the time of measurement, air temperature, and any caffeine consumption in the previous hour. We measured each participant's height (centimeters), weight (kilograms), waist circumference (centimeters), and salt intake from cooked foods and used a pedometer to assess 24-h physical activity. Details on measurement of blood pressure and the other health and sociodemographic factors related to blood pressure are described elsewhere (11) and provided in Tables S1 and S2.

The location of each participant's home was georeferenced using aerial photographs of the study villages from Google Earth (52). We calculated the shortest distance between each participant's home and the closest highway segment to determine the distance from the highway. Smaller roads within villages are narrow and mainly used for walking.

Analysis. We estimated the geometric means and ranges of exposure to $PM_{2.5}$ mass, BC, WSOM, and hopanes by season and age group. Spearman correlation analysis was used to assess collinearity between nonnormally distributed pollution components.

We analyzed the differences in blood pressure associated with 1-ln(μg/m^3) unit increases in pollution exposure using one- and two-pollutant mixed-effects models to determine if observed associations in one-pollutant models were robust to the inclusion of a second pollutant. We only included the first day of pollution exposure in each season because blood pressure was not measured after subsequent second and third days of PM exposure assessment. The two-pollutant models may also help distinguish between the blood pressure effects of combustion vs. noncombustion pollution (BC vs. PM mass), biomass vs. other combustion pollution (WSOM vs. BC), and biomass vs. other nonbiomass pollution (WSOM vs. PM mass).

We used multivariate regression models from our previous study on $PM_{2.5}$ exposure and blood pressure (11) so that any differences in the results could be unambiguously attributed to the difference in pollution variables. The following variables were included in all regression models: age, waist circumference, physical activity (daily number of steps), socioeconomic status, daily salt intake, day of the week and time of day of blood pressure measurement, and ambient air temperature. Passive tobacco smoking, education, caffeine intake, and self-reported health were neither associated with blood pressure at $P < 0.10$ nor did they change the effect of pollution exposure on blood pressure at ≥10%, and were therefore excluded from the final models.

In a second analysis, we allowed the effect of pollution exposure on blood pressure to vary by village using the following model:

$$y_{ifkj} = \mu + \beta_{ifkj} + \zeta_k(\beta_{kj}) + \gamma x_{ifkj} + \eta z_{ifk} + a_{ifk} + h_{fk} + v_k + \varepsilon_{ifkj},$$

where y_{ifkj} is SBP or DBP for individual i in household f in village k at time j, β_{ifkj} is an individual's exposure, $\zeta_k(\beta_{kj})$ is the random slope at the village level, x_{ifkj} are other individual-level covariates that vary seasonally (e.g., ambient air temperature); z_{ifk} are covariates (e.g., socioeconomic status); a_{ifk} is a random intercept; h_{fk} and v_k represent the random effects to account for correlation at the household and village levels, respectively; and e_{ifkj} is the residual.

For sensitivity analyses, we used a scale of the IQR on log-transformed exposures to facilitate comparison between pollutants with different concentration ranges. To assess the validity of the distance from the highway as a proxy for exposure to motor vehicle emissions, we conducted two-sample t tests on geometric mean hopane exposure among women living in the villages nearest to and farthest from the highway. We also conducted a separate analysis excluding women who reported taking antihypertensive medication, all of whom were >50 y old.

All statistical analyses were performed in STATA 11 (StataCorp LP).

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Supporting Information

Baumgartner et al. 10.1073/pnas.1317176111

SI Text

Overview of Optical and Chemical Particulate Matter Exposure Measurements. All air pollution exposure samples were collected on Teflon filters and first analyzed for fine particulate matter ($PM_{2.5}$) mass and black carbon (BC). Portions of each filter were then used for chemical analysis to estimate water-soluble organic carbon (WSOC). We analyzed a subset of PM samples for nonpolar organic hopanes, which are specific markers of motor vehicle emissions in our study setting. More detail about these specific PM components and their measurement is described below.

BC. BC refers to the dark, light-absorbing components of aerosols that contain elemental carbon. Although BC and elemental carbon are often used to describe the same PM fractions, BC is an operationally defined term which describes carbon measured by light absorption. It is therefore different from elemental carbon which is usually measured using thermal–optical methods. BC is emitted from combustion processes, and primary sources include household use of biomass and coal fuels, combustion engines (especially diesel), heavy oil- or coal-fired power stations, and the field burning of agricultural wastes. In addition to climate warming, other regional climate impacts of BC may include increased glacial retreat and changes in precipitation patterns in Asia (1, 2). In health studies, BC is often used as a surrogate for traffic-related PM and has been more strongly associated with a range of cardiovascular and some respiratory outcomes than PM mass in studies conducted in the United States and Europe (3, 4).

Optical Measurement of BC Exposure. BC is defined as the fraction of carbonaceous aerosol absorbing light over a broad region of the visible spectrum. We estimated BC components of total PM mass based on reflectance analysis using an optical transmissometer data acquisition system (SootScan Model OT21; Magee Scientific). This system provides measurements that are highly correlated with concentrations measured using the National Institute for Occupational Safety and Health (NIOSH) thermal–optical method (5). The optical method used in our study both measures and compares the transmission intensity of light at 880 nm passing through an exposed Teflon filter with that of a blank, unexposed filter. The resulting light absorption (ATN) coefficient was computed based on the amount of light transmitted through the exposed filter (I) and the amount transmitted through the blank filter (I_0), where $ATN = 100\ln(I_0/I)$.

We determined BC density (micrograms per square centimeter) by dividing ATN by the specific attenuation coefficient σ_{ATN}, using the relationship, $BC = ATN/\sigma_{ATN}$ (6). BC density was converted to BC exposure concentration for women in our study using the total volume of air (cubic meters) that passed through the exposed Teflon filters during 24-h sampling periods. To account for underestimation of ATN at higher BC concentrations during reflectance analysis (7), we applied an empirical correction factor described elsewhere (8). The instrument was rezeroed with a blank filter between measurements. We obtained the final BC exposure estimate for each participant by averaging five replicate measurements conducted on each filter sample. BC was undetectable on all field blanks.

WSOC and Organic Tracers. Organic carbon can be emitted both from primary emission sources (e.g., biomass combustion, motor vehicles, and industrial sources) and from chemical reactions of gaseous organic precursors in the atmosphere (9). Particulate WSOC accounts for a large proportion of organic carbon in certain settings and may influence the Earth's radiative forcing by acting as cloud condensation nuclei (10). Biomass combustion is a primary source of WSOC in the atmosphere (11, 12). In the absence of emissions from biomass combustion, WSOC is often considered a proxy for secondary organic aerosols due to the highly oxidized nature of water-soluble organic aerosol species (13). Thus, whereas BC serves as a more direct marker of combustion PM, the water-soluble components of PM may be more specific to biomass combustion (14). For example, WSOC accounted for as much as 75% of total PM mass during prescribed biomass burning events in Amazonia (15, 16).

We focused our analysis of source tracers on nonpolar organic hopanes, which are compounds that are present in the lubricating oil of engines (17). Hopanes are relatively involatile and stable compounds that are present in the emissions from mobile sources, fuel oil combustion, and coal combustion (18). In our study site where there is very minimal or no coal or fuel oil combustion, hopanes are tracers for motor vehicle emissions and can be used to distinguish diesel and gasoline engine emissions from other sources of organic carbon (19). We focused our analysis on hopanes as they are more specific and robust markers of motor vehicle emissions than other molecular markers like steranes and polycyclic aromatic hydrocarbons.

To analyze samples and blanks for WSOC, we placed 1.5 cm^3 punches from the Tefon filters in capped conical glass flasks containing 12 mL purified water (Cascada IX; Pall Corp.). We agitated the flasks on a shaker table (120 rpm) at room temperature for 16-h (20, 21). Filter debris and suspended insoluble particles were removed from the water extracts using a polyethersulfone hydrophilic syringe filter (0.45-μm membrane). WSOC was measured with a Shimadzu TOC-V CSH/CSN Total Organic Carbon Analyzer (Shimadzu Corp.) using the nonpurgeable organic carbon method (22). Analytical precision for this method typically falls in the range of 1% to 4% relative SD. The reported WSOC exposures were obtained by averaging the results from three replicates for each filter. The limit of detection for the $TOC\text{-}V_{CPH}$ is 4 μg/L and the SD of the repeated measurements is <1.5% (values provided by manufacturer). All exposure samples were laboratory and field blank subtracted to correct for WSOC contamination of filters, glassware, etc. Blank subtractions were conducted using the average of all field blanks [mean (SD) of blanks = 2.98 (0.71) μg]. Finally, WSOC exposure estimates were multiplied by 2.0 to yield water-soluble organic mass (WSOM) from biomass combustion for statistical analysis, according to Turpin and Lim (23) and Bae et al. (24).

The extraction and analysis methods to quantify individual organic compounds were based on established solvent extraction methods (25). We analyzed samples and blanks for three hopane compounds, specifically 17α(H)-22,29,30-trisnorhopane, 17α(H),21β(H)-hopane, and 17α(H),21β(H)-29-norhopane. Procedures for sample extraction and molecular quantification for the organic tracers are described elsewhere (25) and thus only a brief summary is presented here. The filter samples were spiked with known quantities of isotope-labeled internal standard compounds, then extracted for 15 min with dichloromethane and methanol using an ultrasonic bath. This procedure was repeated three times to ensure adequate recovery of organic carbon mass. The extract was transferred into a round-bottom flask (250 mL) and concentrated to 0.5mL using a rotary evaporator. Suspended particles were removed with a micro-syringe and filter, and the filtrate was collected in a centrifuge tube. To displace the solvent, 5 mL of dichloromethane was added into the centrifuge tube, and the mixture was evaporated to 0.5 mL under a gentle stream of nitrogen gas. We repeated this step twice.

Samples were analyzed using splitless autoinjection into a GC/MS system (GC model 6890 and MSD model 5975; Agilent) equipped with a 30 m × 0.25 mm × 0.25 μm fused-silica capillary column. Along with the samples, six dilutions of authentic quantification standard mixture solutions were also injected and used to build the calibrations curves for each compound. The precision of the spike and standards were used to estimate method precisions because duplicate samples were not available (26). Field blank concentrations of hopanes were below analytical detection limits.

Glassware used during chemical analysis was washed in the ultrasonic bath, rinsed with deionized water, and then baked at 550 °C for at least 6 h before use. The water was deionized and purified in a water purification system (Cascada IX; Pall Corp.). The standard mixture solutions used for calibration curves were obtained from University of Wisconsin-Madison.

Sensitivity Analyses Results. In our analysis comparing hopane exposure for women in the village closest to the highway vs. women in the village farthest from the highway, we excluded 17α(H),21β(H)-29-norhopane due to the large number of exposure samples falling below the limit of detection ($n = 23$ or 32%, of samples from women away from the highway; $n = 5$ or 13% of samples from women near the highway). For women whose exposure was below the limit of detection for the remaining two hopane compounds [$n = 6$ samples or 8% of total samples) for both 17α(H)-22,29,30-trisnorhopane and 17α(H),21β(H)-hopane], we estimated a 99% confidence interval (CI) for each compound's detection limit and then randomly assigned a concentration within this interval to each observation where the measured concentration was below the detection limit. Notably, of the 12 samples below the limit of detection, only one was from a woman in the village near the highway and the remaining samples were from women in the village farthest from the highway.

Geometric mean hopane exposure among women living near the highway was more than twice that of women living away from the highway (Table S4). In winter, geometric mean daily exposure among women near the highway was 6.0 ng/m^3 (95% CI, 4.1 to 8.8) compared with 3.0 ng/m^3 (95% CI, 2.1 to 4.3) for women away from the highway. In summer, average hopane exposure among women near the highway was just slightly lower (5.2 ng/m^3; 95% CI, 3.7 to 7.7) whereas exposure among women living away from the highway decreased considerably (1.9 ng/m^3 (95% CI, 0.9 to 4.0). Hopane exposure was higher in the winter for women in both villages. This may be due to a number of factors including varying environmental conditions (e.g., temperature and humidity) as well as seasonal differences in traffic levels or women's individual behavioral patterns.

As an additional sensitivity analysis, we evaluated the associations of systolic and diastolic blood pressure (SBP and DBP) with exposure to $PM_{2.5}$ mass, BC, and WSOM and expressed the results as the changes in blood pressure associated with a 1-interquartile range (IQR) increase in log-transformed pollutant exposure using one- and two-pollutant multivariate mixed-effects models. We conducted the same multivariate mixed-effects models as those reported in *Materials and Methods, Analysis*.

Supporting our findings in the main text, an IQR increase in ln (BC) exposure had the largest independent effect on both SBP and DBP (Fig. S2). In models with just one pollutant, an IQR increase in ln(BC) was the most strongly associated with higher SBP (3.6 mmHg; 95% CI, 2.0 to 5.2), followed by ln($PM_{2.5}$) (2.8 mmHg; 95% CI, 1.0 to 4.6) and ln(WSOM) (1.8 mmHg; 95% CI, 0.1 to 3.5). The estimated effect of BC on SBP was minimally affected by the inclusion of $PM_{2.5}$ mass or WSOM (<8% change) and it remained statistically significant after adjusting for other pollutants. In contrast, the estimated of $PM_{2.5}$ mass on SBP decreased by 71% and lost statistical significance when BC exposure was added to the model. WSOM had no effect on SBP after other pollutants were included in the model (Fig. S2*A*). We found similar results for DBP (Fig. S2*B*).

We also conducted a separate analysis for younger (25–50 y) vs. older women (>50 y) with the IQR in log-transformed pollution as our pollution measure. Supporting our findings in the main text, BC was more strongly associated with blood pressure than PM mass among both younger and older women. An IQR increase in ln(BC) exposure was associated with a 1.5-mmHg (95% CI, 0 to 3.0) higher SBP in younger women at the sample average, compared with 0.9 mmHg (95% CI, −0.9 to 2.7) for $PM_{2.5}$. Among women >50 y old, an IQR increase in ln(BC) exposure was associated with a 6.2-mmHg (95% CI, 3.4 to 9.0) higher SBP and a 2.4-mmHg (95% CI, 0.9 to 3.9) higher DBP compared with the 5.0-mmHg (95% CI, 1.9 to 8.1) higher SBP and 2.2-mmHg (95% CI, 0.5 to 3.9) higher DBP associated with an IQR increase in ln($PM_{2.5}$) exposure. In both age groups, the estimated effect of BC on blood pressure was minimally affected by the inclusion of $PM_{2.5}$ mass. In contrast, the estimated of $PM_{2.5}$ mass on blood pressure decreased considerably when BC was also included in the model (Fig. S3).

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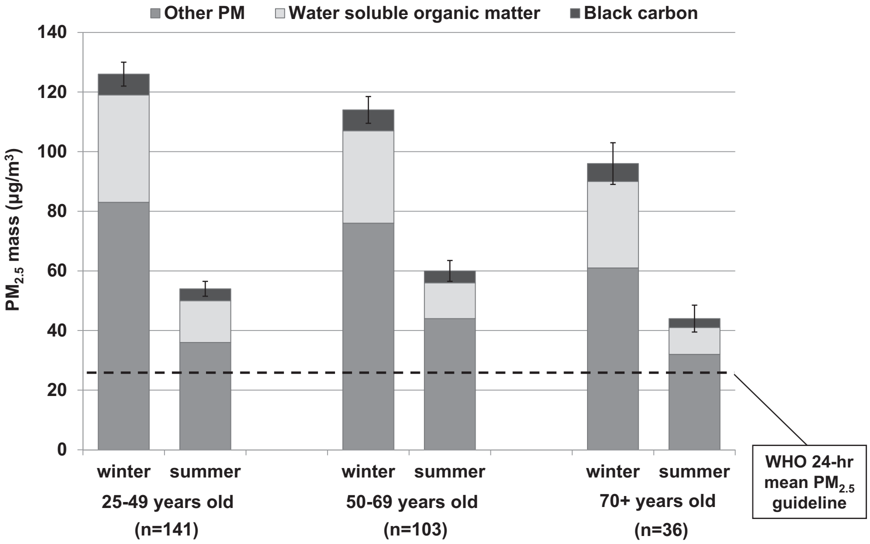

Fig. S1. Geometric mean daily personal exposure to $PM_{2.5}$ mass, BC, and WSOM (micrograms per cubic meter) in rural Chinese women, by season and age. The error bars indicate the 95% CIs for geometric mean $PM_{2.5}$ mass exposure. We used the three age groups presented in the figure because 50-y-old women may reduce their participation in agricultural work and spend more time at home, and 70-y-old women may also participate less often in household tasks like cooking. WHO, World Health Organization.

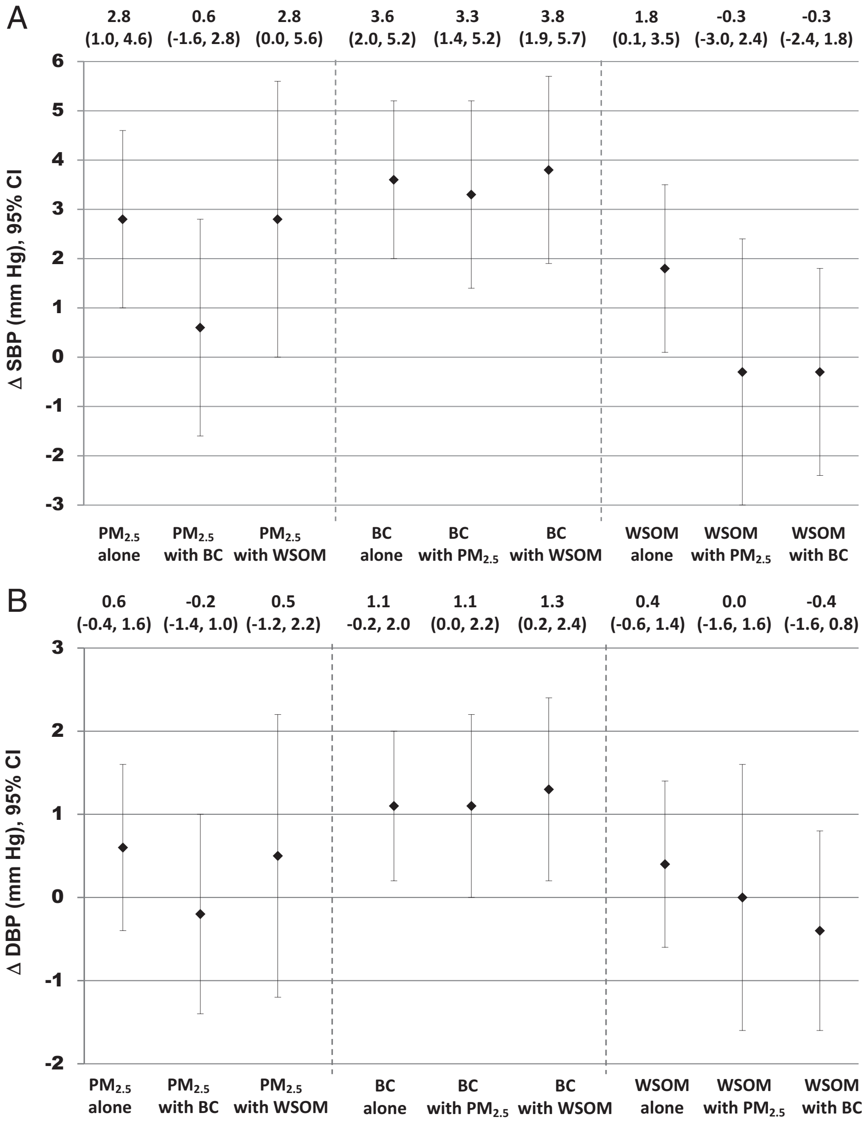

Fig. S2. Associations of IQR increases in log-transformed personal exposure to $PM_{2.5}$ mass, BC, and WSOM on (*A*) SBP and (*B*) DBP using one- and two-pollutant mixed-effects regression models. ΔSBP or ΔDBP represent the difference in SBP or DBP (with 95% CIs) associated with a 1-IQR increase in log-transformed pollutant exposure (equivalent to 44–148 μg/m^3 for $PM_{2.5}$, 3.3–7.4 μg/m^3 for BC, and 11–45 μg/m^3 for WSOM).

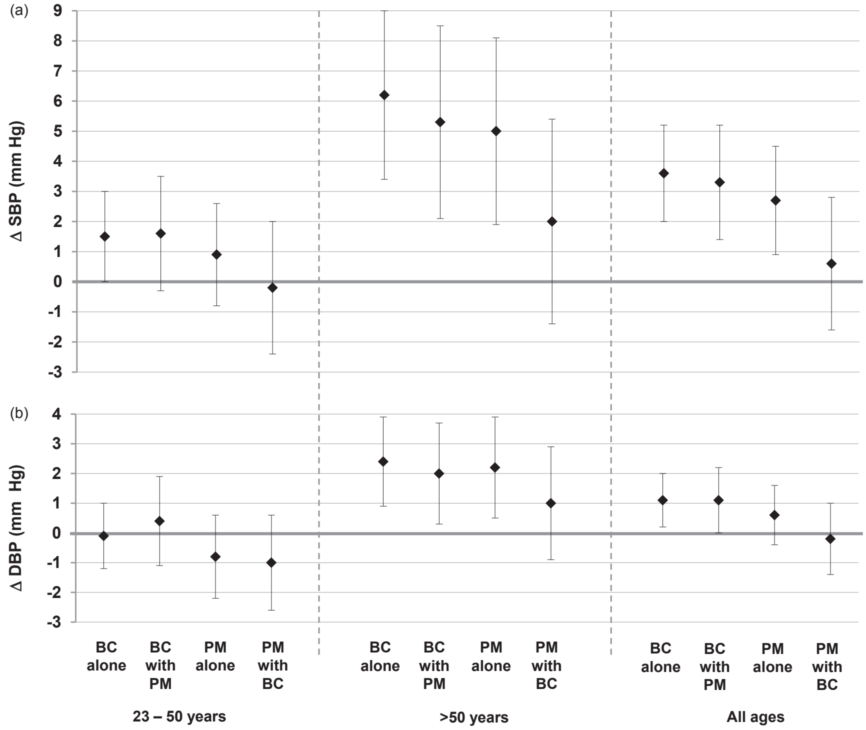

Fig. S3. Associations of IQR increases in log-transformed personal exposure to $PM_{2.5}$ mass and BC on (*A*) SBP and (*B*) DBP using one- and two-pollutant mixed-effects regression models, by age. ΔSBP or ΔDBP represent the difference in SBP or DBP (with 95% CIs) associated with a 1-IQR increase in log-transformed pollutant exposure (equivalent to 44–148 μg/m^3 for $PM_{2.5}$ and 3.3–7.4 μg/m^3 for BC).

Table S1. Demographic and health characteristics of study participants (*n* = 280)

Characteristic	Mean (SD)	Minimum–maximum
Age, y	51.9 (14.3)	25–90
Education, y	5.1 (3.3)	0–13
Waist circumference, cm	81.8 (9.1)	62–119
Body mass index, kg/m^2	22.6 (3.5)	15.6–36.1
Daily salt intake in cooked foods, g	5.6 (3.7)	3.1–26.5
Physical activity: daily no. of steps	7,450 (5,686)	290–41,881
Distance of home from the highway, m	248 (205)	9–1,342
SBP, mmHg	120.0 (19.9)	89.3–200.8
DBP, mmHg	72.4 (10.8)	50.0–116.8

Table S2. Health behaviors and existing cardiovascular conditions among study participants (*n* = 280)

Characteristic	No. (%)
Lives with at least one tobacco smoker	222 (80)
Hypertensive*	37 (13)
Currently taking antihypertensive medication	6 (2)
Physician-diagnosed diabetes	2 (<1)
Self-report of previous cardiovascular event	None

*Defined as SBP ≥ 140 mmHg and/or DBP ≥ 90 mmHg and/or current use of antihypertensive medication.

Table S3. Exposure correlation matrix for personal exposure to $PM_{2.5}$ mass and its components, by season

	Summer				Winter			
Pollutant	$PM_{2.5}$ mass	BC	WSOM	Hopanes*	$PM_{2.5}$ mass	BC	WSOM	Hopanes*
$PM_{2.5}$ mass	1.00				1.00			
BC	0.63	1.00			0.34	1.00		
WSOM	0.72	0.72	1.00		0.60	0.32	1.00	
Hopanes*	−0.65	−0.18	−0.38	1.00	−0.08	0.05	−0.18	1.00

PM, BC, and WSOM exposures are in micrograms per cubic meter, hopane exposure is in nanograms per cubic meter, and the results are for the Spearman rank correlations.
*Analysis of the correlation between hopanes and other pollutants is limited to the subsample of women in the village nearest to and farthest from the highway (*n* = 85 women).

Table S4. Geometric mean BC (μg/m^3) and hopane exposure (ng/m^3) in rural Chinese women, by season and distance from the highway

	Winter			Summer		
Pollutant	Near highway* GM (95% CI)	Away from highway GM (95% CI)	*P* value†	Near highway* GM (95% CI)	Away from highway GM (95% CI)	*P* value†
BC	6.9 (6.3 to 7.7)	5.8 (5.2 to 6.5)	0.01	3.7 (3.5 to 3.9)	4.0 (3.6 to 4.4)	0.15
Hopanes: total‡	6.0 (4.1 to 8.8)	3.0 (2.1 to 4.3)	0.01	5.2 (3.7 to 7.7)	1.9 (0.9 to 4.0)	0.02
17α(H)-22,29,30-trisnorhopane	2.7 (1.9 to 4.0)	0.8 (0.4 to 1.5)	0.02	1.8 (1.2 to 2.8)	0.5 (0.2 to 1.6)	0.28
17α(H),21β(H)-hopane	2.5 (1.2 to 5.4)	1.6 (1.0 to 2.5)	0.008	2.1 (2.2 to 4.5)	0.4 (0.1 to 1.6)	0.03

GM, geometric mean.
*For BC exposure (*n* = 280 women), near-highway exposure is for women in all study villages living in homes within the median distance from the highway (e.g., <208 m) and away-from-highway exposure is for women in homes farther than the median distance. For the subset of women with hopane exposure measurement (*n* = 85 women), near-highway refers to women living in the village closest to the highway (median distance = 76 m) and away-from-highway refers to women in the village farthest from the highway (median distance = 548 m).
†The statistical significance of the difference in geometric mean pollutant exposure between villages, under the null hypothesis that the geometric mean exposures of women in these groups are equal.
‡The sum of the two individual compounds reported here.